

Large amplitude drop shape oscillations

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An experimental study of large amplitude drop shape oscillation has been conducted in immiscible liquids systems and with levitated free liquid drops in air. In liquid-liquid systems the results indicate the existence of familiar characteristics of nonlinear phenomena. The resonance frequency of the fundamental quadrupole mode of stationary, low viscosity Silicone oil drops acoustically levitated in water falls to noticeably lower values as the amplitude of oscillation is increased. A typical, experimentally determined relative frequency decrease of a 0.5 cm^3 drop would be about 10% when the maximum deformed shape is characterized by a major to minor axial ratio of 1.9. On the other hand, no change in the fundamental mode frequency could be detected for 1 mm drops levitated in air. The experimental data for the decay constant of the quadrupole mode of drops immersed in a liquid host indicate a slight increase for larger oscillation amplitudes. A qualitative investigation of the internal fluid flows for such drops has revealed the existence of steady internal circulation within drops oscillating in the fundamental and higher modes. The flow field configuration in the outer host liquid is also significantly altered when the drop oscillation amplitude becomes large.

Introduction

In this paper we report the outcome of a series of experiments aiming at determining the characteristics of large amplitude liquid drops shape oscillations. Two systems have been investigated in the present study: drops held stationary in an immiscible liquid host, and drops freely suspended in air. They have been made accessible to a controlled laboratory study through the technique of acoustic levitation 1-3. Most of the experimental results presented here, however, deal with immiscible liquids systems.

Observations of the variations of the free decay frequency, the fundamental resonance frequency of a forced vibrating drop, the damping constant, and of the time distribution of the prolate and oblate configurations, have been made as functions of the oscillation amplitude. Visualization of the flow fields both inside and outside the oscillating drops suspended in liquid hosts has revealed a gradual appearance of a steady circulation not present for small amplitude oscillations.

Theoretical background

Theoretical analyses of small amplitude drop shape oscillations have been shown to be reasonably successful at describing the observed phenomena 3. Recent linear treatments have included the normal mode approach 4,5, as well as a solution to the initial value problem yielding the small time behavior 6. The analytical solution of the complete nonlinear Navier-Stokes equations have not yet been made available, although results of numerical calculations suggest the appearance of various nonlinear effects as the vibration amplitude of liquid drops suspended in a gaseous medium is increased 7. Among these predictions are a decrease in the free decay frequency and an unbalance in the time distribution of the prolate and oblate configurations characteristic of the fundamental mode.

On the basis of a linear treatment 4,5, the steady-state frequencies for the oscillation modes of a liquid droplet immersed in an immiscible host liquid can be written

$$\omega_L = \omega_L^* - (\alpha/2) \omega_L^{* \frac{1}{2}} + \alpha^2/4 \quad \text{, and} \quad \omega_L^* = [\sigma L(L+1)(L-1)(L+2)/R^3 |L \cdot \rho_o + (L+1) \cdot \rho_i|]^{1/2} \quad (1)$$

The subscripts *i* and *o* refer to the inner and outer fluids respectively. ρ designates the density, ν the dynamic viscosity, σ the interfacial tension, and *L* refers to the mode number. The parameters α and γ are characteristics of the fluids involved. 5

The damping constant for small amplitude oscillations can be written as

$$\tau_L^{-1} = \frac{1}{2} \alpha \omega_L^{* \frac{1}{2}} + \frac{1}{2} \gamma - \frac{1}{2} \alpha^2 \quad . \quad (2)$$

According to these results no dependence on the oscillation amplitude for either the resonance frequency or the damping constant can be obtained, as they are valid only in the limit of small displacements of the drop surface.

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Photographs of experimentally obtained axisymmetric drop shape oscillations in the first three modes are reproduced in figure (1).

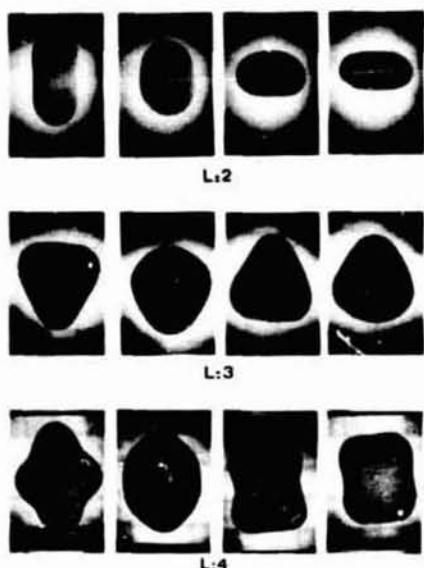


Figure 1 : Experimentally obtained resonance modes for shape oscillation of a 1cm^3 silicone oil drop in water.

Experimental technique

An acoustic standing wave established in a fluid-filled resonant cavity can be used to yield a stable positioning of a fluid sphere having different properties than the cavity fluid. This effect is the result of the action of acoustic radiation pressure forces, and can be obtained in liquids as well as in gases. Figure (2) illustrates such a system. This schematic representation is for a system involving a liquid drop trapped in a sound field established in an immiscible liquid host. In this particular case, the drop liquid has a higher compressibility and density than the host liquid, and is positioned near an acoustic pressure maximum.

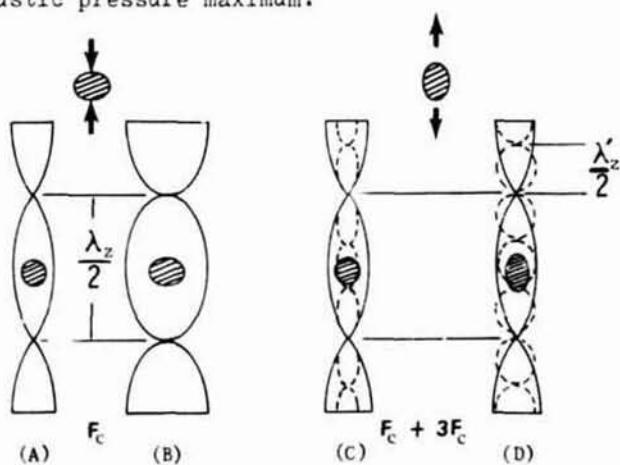


Figure 2 : Schematic representation of a liquid drop trapped in a standing wave. Figure 2b illustrates the action of the acoustic force deforming the drop into an oblate shape. Figure 2d shows the opposite action: the drop is elongated at the poles into a prolate configuration. In this work $f_c = 22\text{ kHz}$.

The acoustic forces will also produce a deformation of the trapped drop when the acoustic pressure is increased (see figure 2a). A low frequency modulation of this force is therefore possible through the modulation of the sound pressure. A steady-state drive of shape oscillations can then be obtained, and the successive drop resonances will be excited as the modulation frequency is varied.

In addition to the deformation shown where the drop is statically distorted into an oblate spheroid, the appropriate tailoring of the acoustic field distribution can provide static distortions into the prolate spheroidal shape. Hence, the opposite drive where the drop is pulled apart at the poles is also available (see figure 2c and 2d).

Figure (3) is a schematic diagram of the experimental apparatus. The necessary electronic instrumentation provides the required voltage drive to a piezoelectric transducer used to excite the acoustic standing waves. An optical detection system allows the monitoring of the shape of the drop. The analog signal thus obtained can be displayed on a CRT screen or can be plotted on a X-Y recorder as a DC signal proportional to the amplitude of oscillation. Resonance curves and free decay traces can be obtained in a straightforward manner. High speed cinefilms also provides the necessary data for the oscillation amplitude and the static equilibrium shapes.

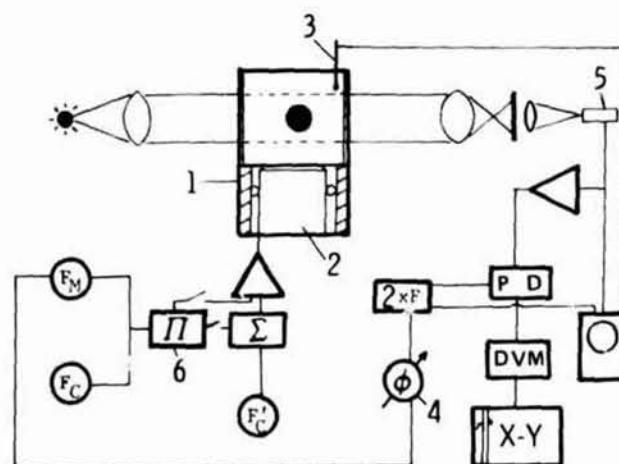


Figure 3 : The experimental apparatus. 1 is the acoustic cell, 2 is the piezoelectric transducer, 3 is a hydrophone, 4 is a variable phase shifter, 5 is a photodetector, 6 is a balanced modulator. The $2xf$ frequency doubler supplies a reference signal with a determined phase relationship with respect to the hydrophone signal.

The voltage applied to the transducer terminals may be written

$$V_T = V_c \sin(2\pi f_c t) \cdot \cos(2\pi f_m t) \quad (3)$$

f_c is the frequency of the high frequency standing wave (22 or 66 kHz in this case), and f_m is the modulation low frequency (from 2 to 15 Hz in this case for 1 cm diameter drops of oil in water). The acoustic pressure is proportional to this voltage for the linear operation of the transducer. The acoustic radiation pressure force is approximately proportional to the square of the acoustic pressure.

$$P_r \sim \langle P_{\text{acous}}^2 \rangle \sim \cos^2(2\pi f_m t) \quad (4)$$

The time average of this force is given by

$$\langle P_r \rangle \sim \langle \frac{1}{2}(1 + \cos 4\pi f_m t) \rangle \quad (5)$$

This includes a steady-state as well as a slowly varying force component at the frequency $2f_m$. The steady-state component induces a steady deformation of the drop shape.

Free decay frequency

Measurements of the free decay frequency in the immiscible liquid system as a function

of the initial oscillation amplitude were carried out by first exciting the drop into its first resonant mode. The modulated acoustic drive was then turned off and the decay phase observed. The f_1 decay frequency was measured from both the oscilloscope traces and high speed motion picture films. The experimental uncertainty was within $\pm 1\%$.

In the axisymmetric case, and with the linear approximation, one might describe the deformation with the parameter

$$x(\theta, t) = r(\theta, t) - R_0 \quad , \quad (6)$$

where $r(\theta, t)$ is the expression for the boundary of the drop, and R_0 is the radius of a sphere of equal volume. A linear expansion would yield

$$x(\theta, t) \approx \sum_{n=2}^{\infty} \left[x_n^{\text{static}} + x_n \cos(4\pi f_m t + \phi_n) \right] P_n(\cos\theta) \quad . \quad (7)$$

$P_n(\cos\theta)$ is a Legendre polynomial of order n . In this particular case where only the fundamental mode is excited, and for a reasonably high Q system ($Q=15$), one has the approximations

$$x_2 \gg x_n \gg x_n^{\text{static}} \quad (8)$$

x_n^{static} is the distortion produced by the static component of the acoustic force.

During free decay the drop deformation may be expressed as

$$x(\theta, t) \approx \sum_{n=2}^{\infty} \left[x_n^{\text{static}} \cos(2f'_n t + \phi'_n) \exp(-b'_n t) + x_n \cos(2f''_n t + \phi''_n) \exp(-b''_n t) \right] P_n(\cos\theta) \quad . \quad (9)$$

The term with the single prime refers to the decay of the static deformation, the double primed one to the decay of the oscillatory motion initially driven at the resonance frequency of the fundamental mode. In this particular case the time dependence of f'_n , f''_n , b'_n , and b''_n has been neglected. This has been justified by the experimental evidence.

Figure (4) reproduces some of the experimental results. The free decay frequency variations with the maximum oscillation amplitude prior to the decay phase are shown for a 0.5 and a 1 cm³ drop of Silicone oil/CCl₄ mixture made almost neutrally buoyant with distilled water. A steady decrease can be observed with increasingly larger initial oscillatory motion.

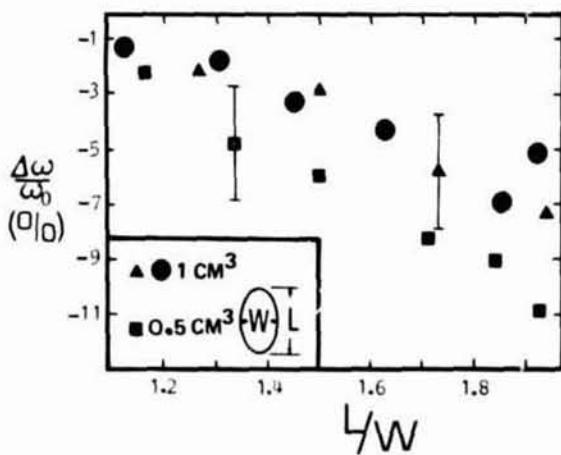


Figure 4 : Relative change in the free decay frequency as a function of the initial oscillation amplitude. The ratio L/W refer to the maximum deformation in the prolate shape during the steady-state forced oscillations prior to the free decay phase.

Driven oscillations

The amplitude dependence of the fundamental resonance frequency for forced steady-state oscillations has been investigated both in the immiscible liquids system, and with drops suspended in air. The results are qualitatively different for the two systems, although the measurements made for a levitated drop in air are not as precise, and for significantly distorted drops due to the gravitational field.

Figure (5) is a plot of the relative frequency shift as a function of the oscillation amplitude for a 1 cm^3 drop of Silicone oil/CCl₄ mixture in distilled water. Qualitatively different results are obtained when the two opposite driving mechanisms are used. The oblate-biased mechanism is based on a compression of the drop at the poles, while the prolate-biased drive consists in elongating the drop at the poles. The source of such a discrepancy has not yet been totally elucidated, but there are some indications that the interference of the acoustic fields on the drop motion might play an important role.

Figure (6) reproduces the results of measurements taken for drops of a mixture of glycerin and distilled water suspended in air. The equilibrium shape of the drops is oblate as indicated by the parameter W/L . No change in the fundamental mode resonance frequency can yet be resolved within the experimental uncertainty ($\pm 5\%$). These results have been obtained with an oblate-biased mechanism.

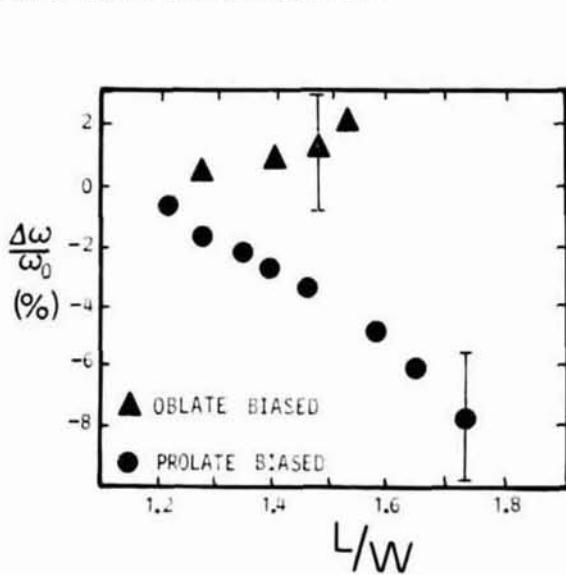


Figure 5: Amplitude dependence of the fundamental mode for forced oscillations for a liquid drop in H₂O.

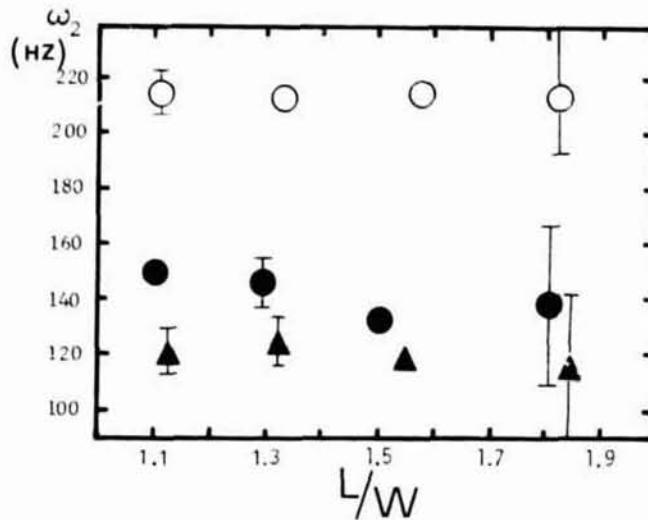


Figure 6: Amplitude dependence of the fundamental mode frequency for a drop levitated in air. $\bigcirc R=0.097 \text{ cm}$, $\bullet R=0.125 \text{ cm}$, $\blacktriangle R=0.151 \text{ cm}$. The static shape is oblate. $(W/L) \approx 1.2$.

Measurement of the dissipation rate of liquid drops oscillating in an immiscible liquid host has revealed a single value for the free decay constant for each initial oscillation amplitude. Figure (7) gives an example of such a decay process. The decrease to zero deformation is strikingly exponential, and is at a constant rate. This rate appears to vary for different initial oscillation amplitude, with a tendency for higher values for larger amplitudes. Figure (8) shows such results.

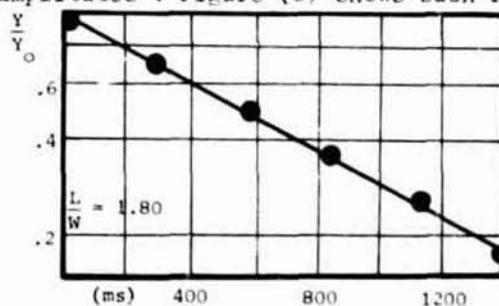


Figure 7: Time dependence of drop oscillation amplitude.

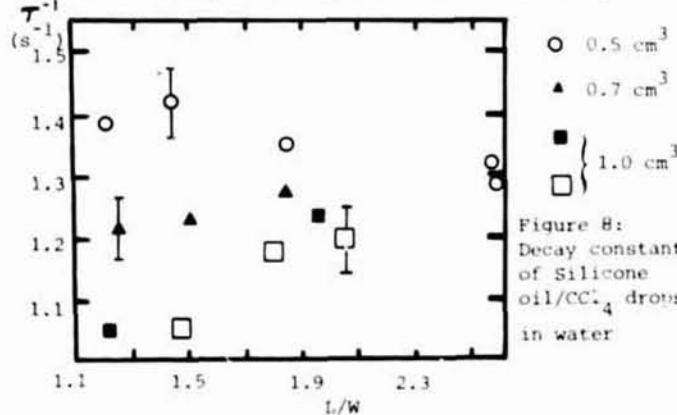


Figure 8: Decay constant of Silicone oil/CCl₄ drops in water

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Time distribution of the prolate and oblate configurations

Immiscible liquids systems results suggest that the duration of the prolate configuration increases with the oscillation amplitude. Table (I) reproduces the results of measurements performed on drops with an oblate static equilibrium shape. The duration of the prolate configuration definitely shows an increase for larger amplitudes. Such an increase is also obtained when a static prolate shape is used.

Cycle number after termination of the steady drive	MAXIMUM OBLATE DISTORTION DURING OSCILLATIONS			
	W/L = 1.75		W/L = 1.31	
	T_{oblate}	$T_{prolate}$	T_{oblate}	$T_{prolate}$
	T_{cycle}	T_{cycle}	T_{cycle}	T_{cycle}
1	0.51	0.49	0.51	0.49
2	0.53	0.47	0.53	0.47
3	0.54	0.46	0.55	0.45
4	0.57	0.43	0.60	0.40
5	0.60	0.40	0.64	0.36

Table I: Duration of the oblate and prolate cycles during the free decay of shape oscillations initially forced by acoustic forces. The static equilibrium shape is oblate. Larger amplitude oscillations increase the duration of the prolate phase.

Fluid flow fields

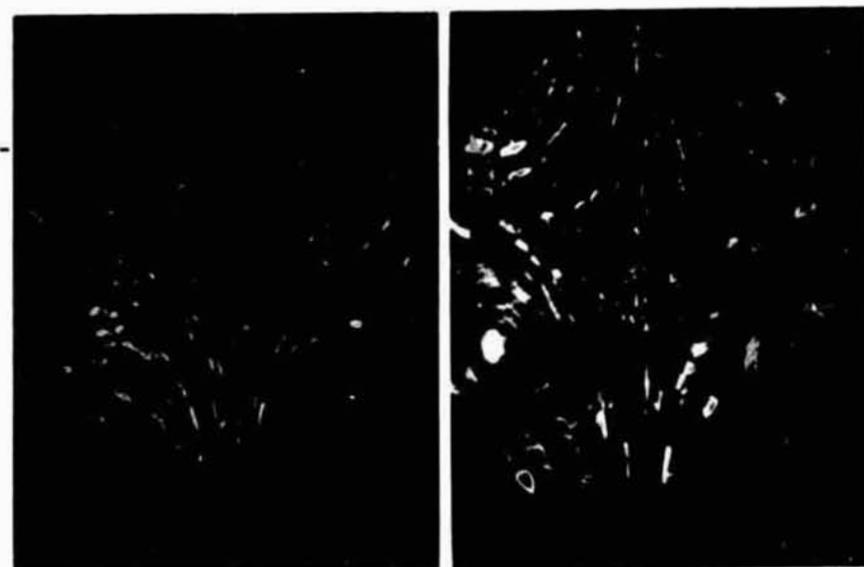
The visualization of the fluid flow fields revealed by suspended dye particles appropriately illuminated, has shown the gradual appearance of a steady circulatory motion superposed upon the oscillatory motion induced by the drop shape oscillations in liquid-liquid systems. Figure (9) shows photographs of such flow fields for small and large amplitude for both inner and outer liquids.



Figure 9: A. Streak patterns of suspended dye particles in a Silicone oil drop undergoing small amplitude oscillation in the L=2 mode. The drop is levitated in distilled water. ($\Delta R/R \leq 0.05$) B. Streak patterns for the same drop oscillating at higher amplitude. ($\Delta R/R \approx 0.1$).

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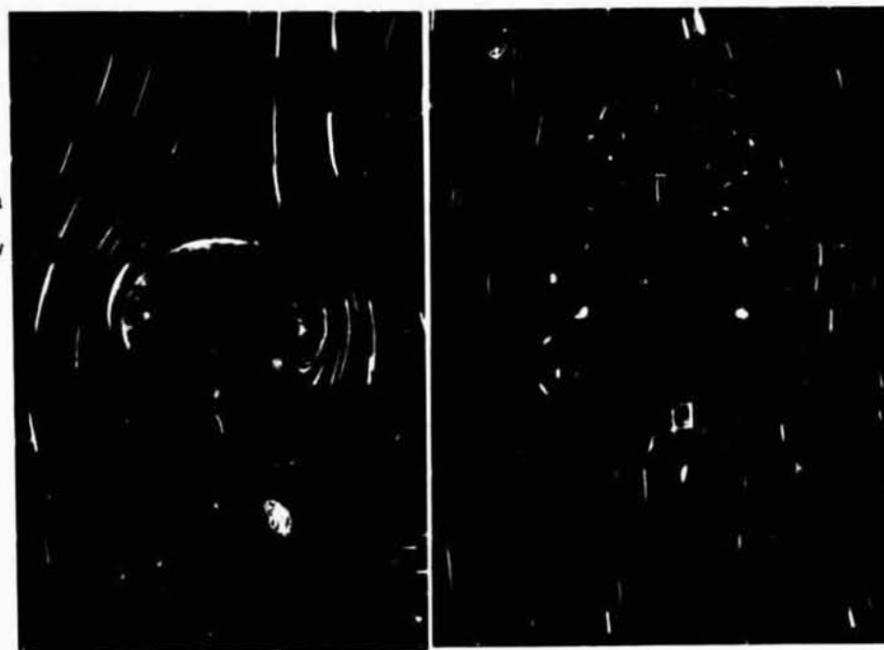
Figure 9: C. Same drop as in A and B, but the oscillations are in the L=3 mode at small amplitude.
D. L=3 oscillations at large amplitude ($\Delta R/R \approx 0.10$).



C

D

Figure 9: E. Flow pattern in the outer fluid with a stationary and still drop. The flow around the drop is caused by acoustic streaming, and is characterized by a Reynolds number of 5.
F. Flow pattern in the outer fluid for a drop oscillating in the L=2 mode at small amplitude.



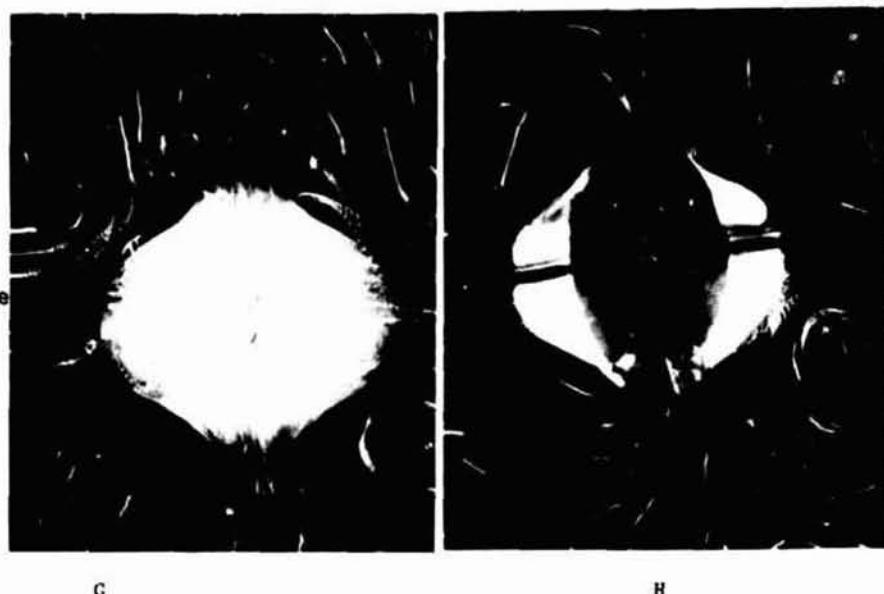
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Figure 9: G. and H.

Flow pattern in the outer fluid with a drop oscillating in the $L=2$ mode at large amplitude.



G

H

Conclusion

The experimental observations gathered so far point to certain definite nonlinear characteristics of liquid-liquid systems. Both inertial and viscous effects have been shown to exist, and these must play an important role in causing these phenomena. The absence of theoretical information, however, has not allowed a further analysis.

The techniques using acoustic radiation pressure forces have been shown to allow the controlled experimental study of single drop phenomena. One must exercise caution, however, when interpreting the observations based on such a technique because of the unavoidable interaction between the acoustic fields and the drop motion.

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